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RESEARCH MEMORANDUM

VARIATION OF PRESSURE LIMITS OF FLAME PROPAGATION WITH TUBE DIAMETER FOR VARIOUS ISOOCTANE-OXYGEN-NITROGEN MIXTURES

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NATIONAL ADVISORY COMMITTEE FOR AERONAUTICS

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SUMMARY

The change in the pressure limits of flame propagation with tube diameter for various isooctane-oxygen-nitrogen mixtures was studied; the effects of oxygen concentration upon the pressure limits and concentration limits of flame propagation were also investigated.

Low-pressure propagation limits were measured for quiescent isooctane-oxygen-nitrogen mixtures in cylindrical glass tubes with inside diameters of 16, 22, 28, and 38 millimeters. Oxygen-nitrogen atmospheres containing 15.0 to 49.6 percent by volume oxygen were used.

The data showed that, under the conditions of the experiments, no flame propagation is possible for isooctane in oxygen-nitrogen mixtures containing less than 11 to 12 percent oxygen.

The experiments gave quenching distances in terms of critical diameter for flame propagation. For approximately stoichiometric isooctane-air mixtures, critical diameter was found to be proportional to the pressure raised to the -0.91 power. The critical tube diameters were on the average 1.25 times as large as quenching distances obtained in connection with ignition-energy experiments.

For experiments in various oxygen-nitrogen atmospheres, the logarithms of the maximum flame speeds for isooctane were proportional to the critical diameters.

INTRODUCTION

The study of combustion processes and their relations to aircraft engines can be approached in two general ways: (a) the study of combustion in the engine itself; and (b) the study of combustion processes in idealized systems. A part of the program in the second category is being carried out at the NACA Lewis laboratory and is considered herein. The eventual aim of all such studies is understanding of the complex combustion process as it occurs in practical applications.

The effects of various proportions of oxygen and inert gases on the combustion properties of hydrocarbon fuels have been studied by a number of investigators. However, most of these studies have involved gaseous fuels. For example, Blanc, Guest, Lewis, and von Elbe studied the effect of oxygen concentration on flammability limits, minimum ignition energies, and quenching distances of methane, ethane, and propane (reference 1). Badin, Stuart, and Pease studied the effect on flame speeds of butadiene (reference 2). In the case of liquid hydrocarbon fuels, a recent investigation of the minimum ignition energies of isooctane-oxygen-nitrogen mixtures has been made by Experiment, Inc., commercial laboratory, under NACA contract.

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A research program on the combustion properties of isooctane-oxygen-nitrogen mixtures, including flame-speed measurements, (unpublished), is also in progress at the NACA Lewis laboratory. The effects of the oxygen fraction $\frac{O_2}{O_2 + N_2}$ on the pressure-flammability limits and concentration-flammability limits of these mixtures is considered herein. Pressure-flammability limit curves were determined in four glass flame tubes of 16 to 38 millimeters inside diameter with six oxygen-nitrogen atmospheres containing 15.0 to 49.6 percent oxygen by volume. From these data, quenching distances were obtained in terms of the critical tube diameters for flame propagation.

EXPERIMENTAL DETAILS

Apparatus. - In this investigation, the tube method of determining pressure-flammability limits was used (references 3 and 4). Special modifications of the basic apparatus were made to permit the study of liquid fuels. The equipment is illustrated in figure 1.

The fuel metering, mixing, and storing apparatus consisted of a 45-liter galvanized steel storage tank with sealed stirrer A, fuel capsule B, air inlet I, and precision manometer C. These components were mounted within a glass-walled tank of ethylene glycol, which served as a constant-temperature bath. The bath temperature was thermostatically controlled to a preset temperature of 58 $\pm$0.5° C.

The test section consisted of a series of interchangeable glass flame tubes. The four flame tubes used in this investigation were 4 feet long and had inside diameters of 38, 28, 22, and 16 millimeters. Each tube was sealed to an ignition section 90 millimeters in diameter and 25 centimeters long. The flame tubes were connected to a 47-liter plenum chamber through a spherical glass joint and a 25-millimeter-bore stopcock. In order to prevent condensation of liquid fuel from the combustible mixtures, the flame tube and the ignition section were enclosed by a cylindrical resistance-wound furnace. Three separate windings were calibrated to give a uniform temperature of 58° C throughout the length of the flame tube. The furnace was provided with a longitudinal slit 1/2 inch wide for visual observation of the flames.

The isooctane-oxygen-nitrogen mixtures were ignited by passing a rapid capacitance spark discharge across pointed stainless-steel electrodes. The maximum voltage across the gap was 30,000 volts.

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<u>Preparation of isooctane-oxygen-nitrogen mixtures.</u> - The oxygen-nitrogen atmospheres used were obtained commercially in cylinders at 2200 pounds per square inch gage except in the case of air. The artificial mixtures contained 15.0, 25.0, 29.4, 34.7, and 49.6 percent oxygen by volume to within ±0.1 percent.

The isooctane used was commercially available knock-rating reference fuel with a purity of 99.6 mole percent. When a combustible mixture was prepared, the fuel mixture and the storage tank were first evacuated. Isooctane vapor was then expanded into the tank from the fuel capsule (see fig. 1). The partial pressure of the isooctane was read on the precision manometer by means of a cathetometer accurate to ±0.05 millimeter. The desired oxygen-nitrogen atmosphere was passed through Anhydrone to remove water and Ascarite to remove carbon dioxide, and then admitted to the tank to form the desired isooctane-oxygen-nitrogen mixture as calculated on the basis of the ideal gas law. In order to insure homogeneity, the mixture was stirred by a vane-type stirrer sealed into the tank. Once a combustible mixture had been prepared as outlined, leaner mixtures could be made from it by successive dilutions with the oxygen-nitrogen mixture.

PROCEDURE

The method of determining pressure-flammability limits was basically the same as that reported in reference 4.

For a given combustible mixture, the pressure was found at which a flame, propagating upward in the ignition section, was just extinguished at the mouth of the narrower flame tube. In many of the experiments, extinction of the flame was observed visually. However, for some of the combustible mixtures with high $\frac{O_2}{O_2 + N_2}$ ratios, the flame moved so rapidly that it was impossible to determine visually whether it propagated the length of the flame tube or was extinguished at the mouth. In these cases, a thermocouple was used to detect the passage of the flame. The thermocouple leads were admitted through the top of the flame tube and the hot junction was positioned about 1/2 inch above the point where the flame tube widened into the ignition section. The thermocouple was 40-gage chromel-alumel wire, connected by 36-gage ceramic-coated copper leads to a rapid-response recording potentiometer.

The operation of this flame detector was checked with flames on which visual observations could be made, and good agreement was obtained between the two types of observation. The location chosen for the thermocouple junction proved to be such that the potentiometer did not register a temperature increase if the flame was extinguished at the mouth of the flame tube. If the flame progressed farther, a temperature increase was recorded by the potentiometer. It was found in this investigation, as in the work reported by reference 4, that these slower flames on which visual observations could be made either propagated the entire length of the tube or were extinguished at the mouth of the tube. The pressure difference between propagation and extinction was very small, only 1 or 2 millimeters of mercury. If the flame propagated, it, of course, caused the potentiometer to register a temperature increase as it passed the thermocouple position. Consequently, the pressure limit could be established in terms of potentiometer scale deflection just as easily and precisely as by visual observation. In addition, it was found that the presence of the thermocouple and leads inside the flame tube had no effect on the pressure limit.

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In the case of the very rapid flames with which the thermocouple system was designed to be used, the demarcation (in terms of pressure) between propagation and extinction of the flame was not so sharp as it was for slower flames. It was found that, as flames were observed at successively lower pressures, the potentiometer at first experienced a full-scale deflection with the passage of each flame. Finally, a test pressure was reached for which a full-scale deflection was not obtained, and subsequent tests at lower and lower test pressures showed smaller and smaller potentiometer deflections. Because of this behavior, the pressure limit for propagation of a fast flame was determined by making a plot of scale deflection against test pressure, and extrapolating the resulting straight line to zero scale deflection. The pressure corresponding to zero deflection was reported as the pressure limit and was checked by making an additional test at a pressure 1 millimeter of mercury lower. Zero scale deflection was invariably obtained. The precision of this pressure limit was ±2 millimeters of mercury.

The pressure at which the first potentiometer deflection less than full-scale occurred was 5 to 10 millimeters of mercury greater than the reported pressure limit. Thus the pressure range of uncertain flame propagation was much greater than the 1- or 2-millimeter of mercury difference in the pressures of propagation and extinction for slow flames. However, it is believed that use of the procedure described herein led to pressure limits for the rapid flames that were consistent with those obtained visually for slower flames.

A given pressure-limit curve obtained by measuring the limits for a series of mixtures could be reproduced on subsequent days with fresh mixtures. The results of check runs are shown as tailed points in figures 2(c) to 2(e).

RESULTS AND DISCUSSION

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Curves of pressure limit against percent stoichiometric isooctane for mixtures of isooctane in an atmosphere of 15 percent oxygen and 85 percent nitrogen in tubes of 38, 28, 22, and 16 millimeters inside diameter are presented in figure 2(a). Figures 2(b) to 2(f) show similar data for isooctane in oxygen-nitrogen atmospheres containing 20.9, 25, 29.4, 34.7, and 49.6 volume percent oxygen. For a given oxygen concentration, the general effect of decreasing tube diameter was to raise the minimum pressure for flame propagation and to narrow the concentration range of flammability, as has been reported previously (references 4 and 5). In general, the curves show irregular lobes and cross-overs on the rich side of stoichiometric (except in the case of the curves for the 15 percent oxygen and 85 percent nitrogen atmosphere), while on the lean side of stoichiometric they are regular. This behavior has been noted in other cases (references 3 to 6). The lobes that appear on the rich side of stoichiometric may be indicative of the presence of cool flames as described in reference 3. In the present investigation, however, capacitance sparks were used, which according to reference 3 are incapable of igniting cool flames.

For 80 to 200 percent stoichiometric isooctane in oxygen-nitrogen atmospheres containing more than 25 percent oxygen and in the flame tubes of 16 and 22 millimeters diameter, the flames propagated so rapidly that it was impossible to decide visually how far they traveled. It was therefore necessary to determine the pressure limit by the thermocouple technique described previously. In several instances with the atmosphere containing 49.6 percent oxygen and 50.4 percent nitrogen, shattering detonations occurred at pressures as low as 25 millimeters of mercury. It is believed that the high flame speeds and detonations, with the consequent effects upon the pressure-limit data, were due in part to the nature of the apparatus, particularly the relatively constricted stopcock (25-mm bore) that connected the test section and the plenum chamber.

The effects of variation in the oxygen concentration of oxygen-nitrogen atmospheres are shown more graphically in figures 3 and 4. For a given tube diameter, decreasing the oxygen concentration raised the minimum pressure for flame propagation and narrowed the concentration range of flammability (figs. 3 and 4). It will be noted from figure 2 that both the lean and the rich sides of the limit curves become substantially vertical at a pressure of approximately 250 millimeters of mercury. Figure 3 shows a plot of flammability range at this pressure against the volume percent of oxygen in the oxygen-nitrogen atmosphere for tubes of 16, 22, 28, and 38-millimeter inside diameter. The flammability range is defined as the rich limit for propagation (in percent stoichiometric) minus the lean limit (in percent stoichiometric). For the range of tube diameters considered, figure 3 indicates that the flammability range for isooctane becomes zero (no flame propagation possible) in oxygen-nitrogen atmospheres containing 11 to 12 percent oxygen by volume. This conclusion is supported by the fact that no propagation could be obtained in 28- and 38-millimeter diameter tubes in a mixture of 9.4 percent oxygen and 90.6 percent nitrogen. It should be emphasized that this limiting oxygen concentration applies only to the conditions of this experiment. In particular, a marked change in the temperature of the experiment would be expected to change this result. It is of interest to note that zero flame speed has been predicted for isooctane in an oxygen-nitrogen atmosphere containing less than 12 percent oxygen (unpublished NACA data).

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The effect of oxygen concentration in the oxygen-nitrogen atmosphere on the minimum pressure for propagation of flame in isooctane-oxygen-nitrogen mixtures is shown in figure 4 for tubes of 16, 22, 28, and 38-millimeter diameter.

Previous work at this laboratory on the pressure-flammability limits of propane-air mixtures (reference 4) showed that data such as those of figure 2 yield a critical diameter for flame propagation. This critical diameter was shown to be a quenching distance, just as the minimum slit width for flashback of a Bunsen flame was shown to be a quenching distance by Friedman and Johnston (reference 7). Figure 5 compares the pressure dependencies of critical diameter, obtained from the present work, and of minimum quenching distance of isooctane-air mixtures. The minimum quenching distances were obtained in connection with ignition-energy measurements made by Experiment, Inc., commercial laboratory, on NACA contract. The critical diameters are seen to depend upon pressure raised to the -0.91 power determined by calculating the slope of the line; the quenching distances also depend upon pressure raised to the -0.91 power. The ratio of critical diameter to quenching distance is 1.25. This value may be compared with previously reported ratios of 1.35 (reference 7) and 1.43 (reference 4) for propane-air mixtures. Figure 5 includes some of the data of references 1 and 7 on propane-air mixtures, for comparative purposes.

As a result of previous work, a simple inverse relation has been found between flame speed and critical diameter or quenching distance for propane-air mixtures (reference 4). The relation held for varying propane concentration (constant pressure and mixture temperature) and for varying mixture temperature (constant pressure and propane concentration). It was found possible to justify the inverse relation on a theoretical basis (reference 8). The present work provides an opportunity to test the relation between flame speed and critical diameter at various concentrations of oxygen in the oxygen-nitrogen mixture. Figure 6 shows the logarithm of the maximum flame speed for isooctane in a given oxygen-nitrogen atmosphere plotted against the critical diameter under the same conditions. The oxygen concentration in the oxygen-nitrogen atmosphere of the mixture is noted beside each point. Pressure in all cases was 1 atmosphere, mixture temperature was 58° C, and isooctane concentration was 105 percent stoichiometric. The flame speeds are from unpublished data; the critical diameters are from the present work, extrapolated to 1 atmosphere pressure. The good degree of correlation shown by figure 6 indicates that, for the conditions under which the data are plotted, flame speed is related to critical diameter in the manner shown by the following expression:

$$\mu_f \propto e^{-cd_1} \qquad (1)$$

where

μ_f	flame speed
c	constant
d_1	critical diameter

instead of being related to the simple reciprocal of the critical diameter. The reasons for the changed relation between flame speed and critical diameter may lie in the variation with oxygen-nitrogen ratio of some of the factors involved. The theory of reference 8 may prove capable of predicting relation (1) as it predicted the reciprocal relation between flame speed and critical diameter in previous cases. A test cannot be made until suitable calculations of adiabatic flame temperatures and equilibrium concentrations of free radicals, for isooctane in various oxygen-nitrogen atmospheres, are used to evaluate the theoretical expressions.

SUMMARY OF RESULTS

From the investigation of the variation of the pressure limits of flame propagation with tube diameter for various isooctane-oxygen-nitrogen mixtures, the following results were obtained:

1. For a given oxygen-nitrogen atmosphere, decreasing tube diameter raised the minimum pressure for flame propagation and narrowed the concentration-range of flammability. For a given tube diameter, decreasing oxygen concentration in the oxygen-nitrogen atmosphere produced the same effects.

2. For the range of tube diameters studied, the flammability range for isooctane became zero (no flame propagation possible) in oxygen-nitrogen atmospheres containing 11 to 12 percent oxygen by volume.

3. The data obtained could be interpreted in terms of critical diameter for flame propagation. The minimum critical diameter was found to depend upon the pressure raised to the -0.91 power. The ratio of critical diameter to quenching distance obtained in connection with ignition energy experiments was found to be 1.25.

4. Flame speed of isooctane in various oxygen-nitrogen atmospheres, for 105 percent stoichiometric isooctane, 1 atmosphere pressure, and 58° C mixture temperature, was found to be related to the negative exponential critical diameter.

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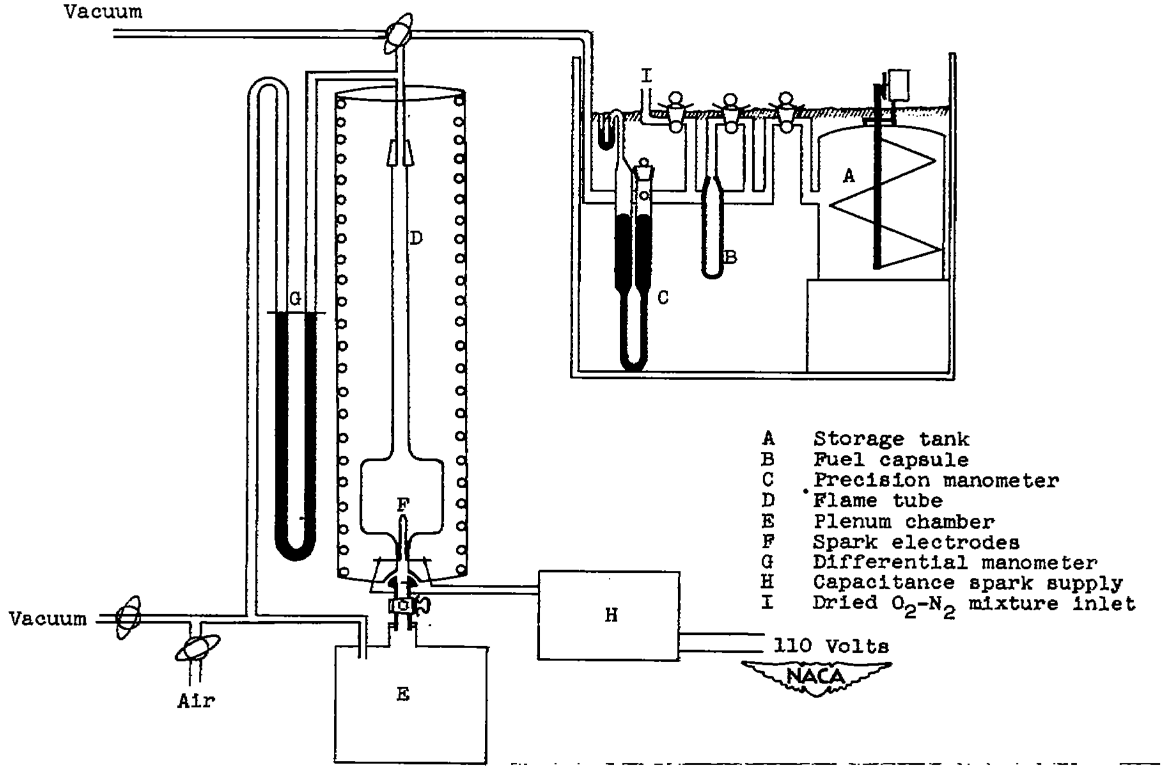

Figure 1. - Apparatus for determination of flammability limit.

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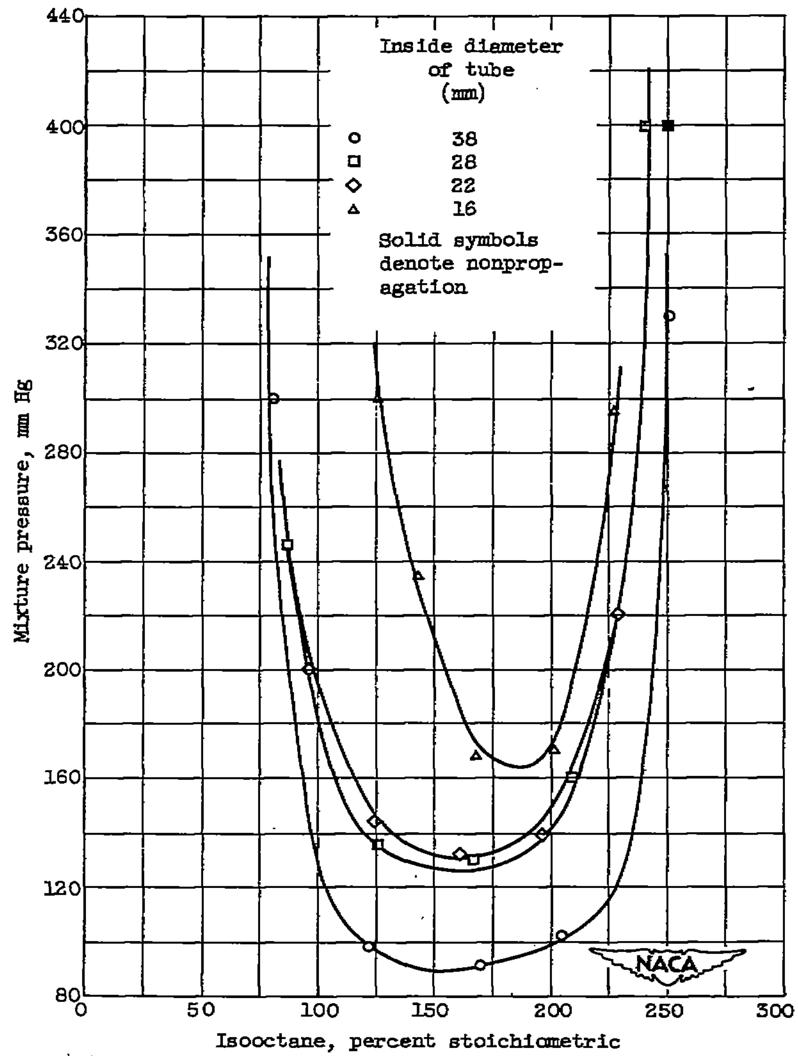

(a) Oxygen fraction $\frac{O_2}{O_2 + N_2}$, 0.15.

Figure 2. - Effect of tube diameter on pressure limit.

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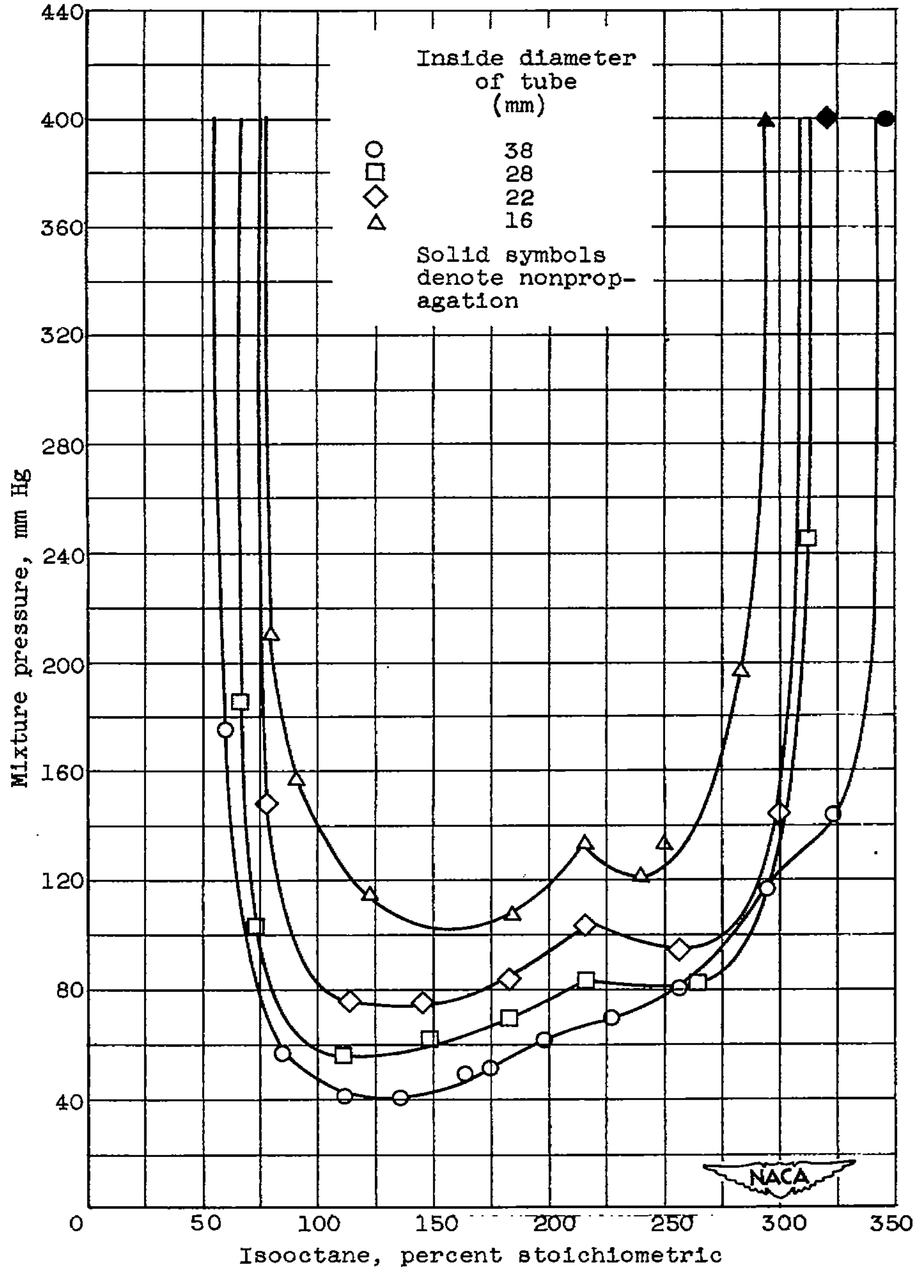

(b) Oxygen fraction $\frac{O_2}{O_2 + N_2}$, 0.21.

Figure 2. - Continued. Effect of tube diameter on pressure limit.

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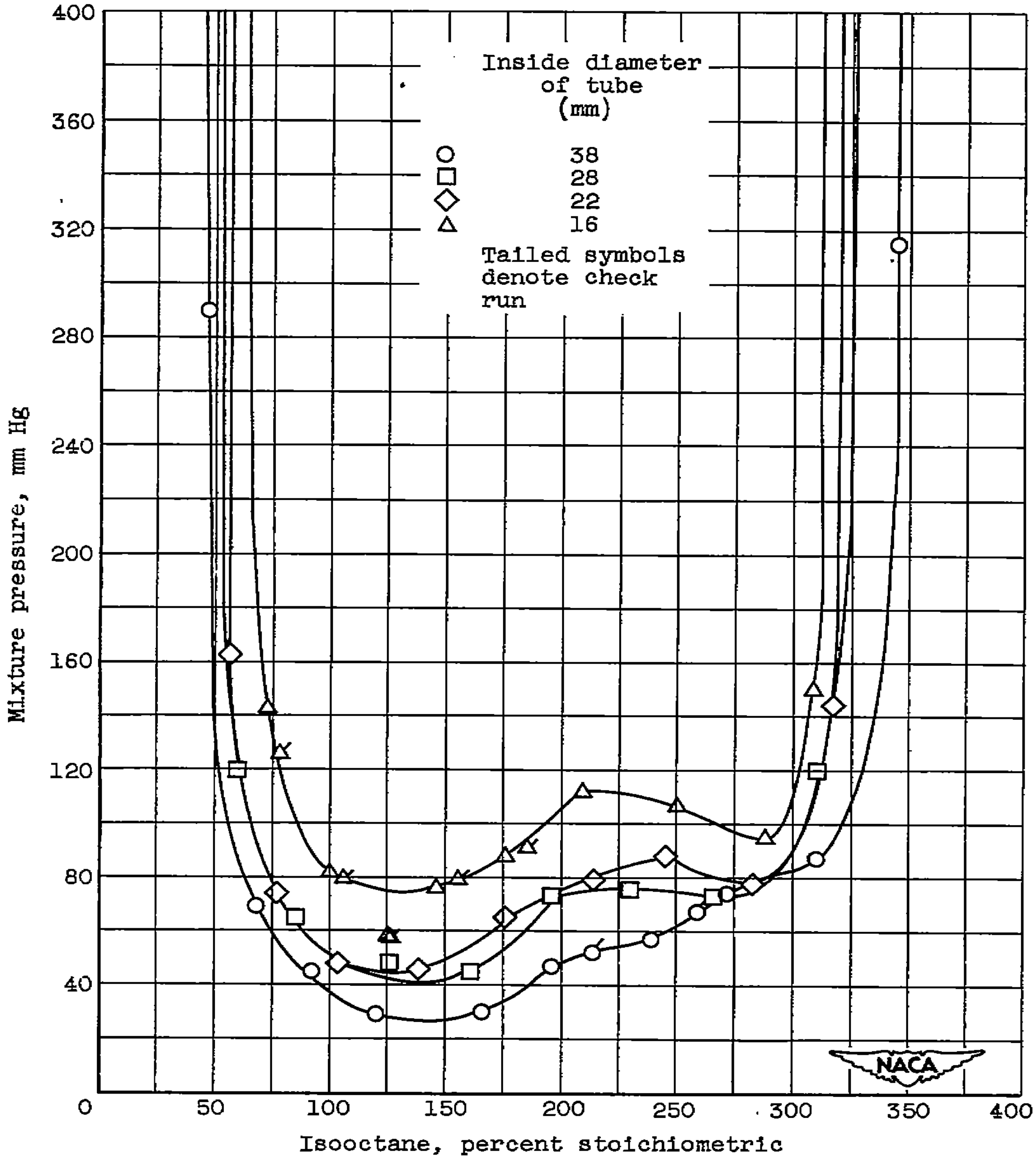

(c) Oxygen fraction $\frac{O_2}{O_2 + N_2}$, 0.25.

Figure 2. - Continued. Effect of tube diameter on pressure limit.

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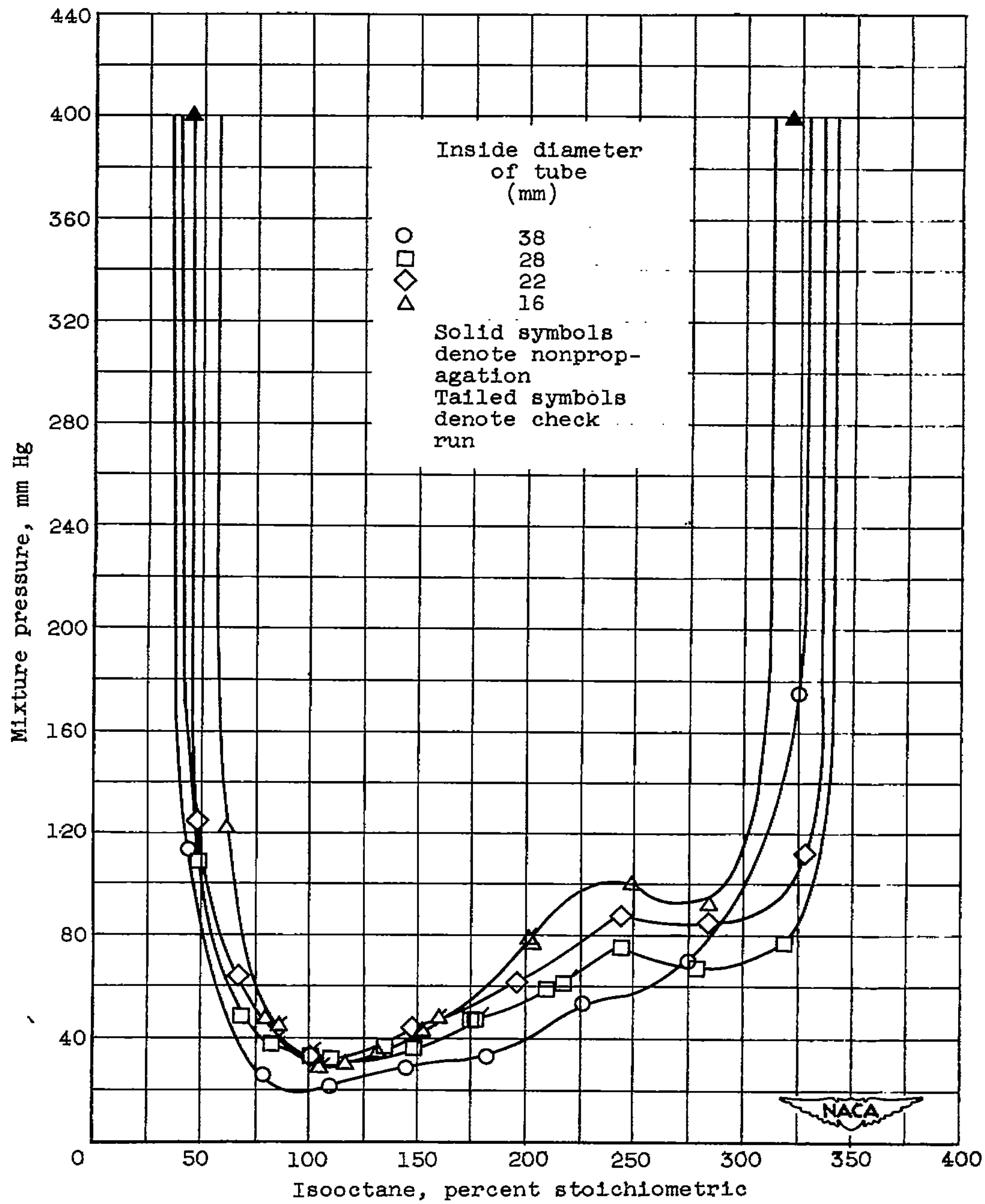

(d) Oxygen fraction $\frac{O_2}{O_2 + N_2}$, 0.294.

Figure 2. - Continued. Effect of tube diameter on pressure limit.

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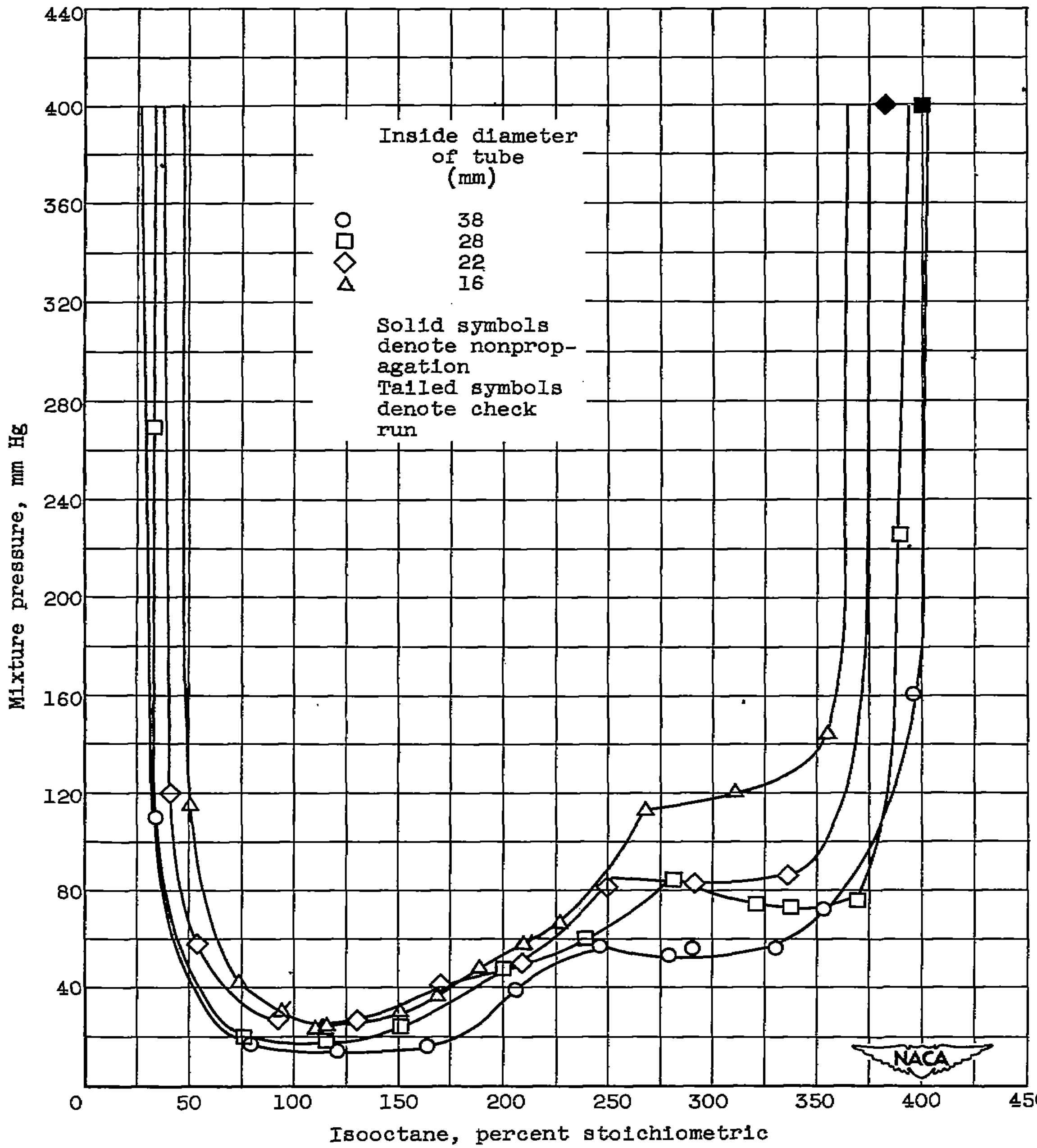
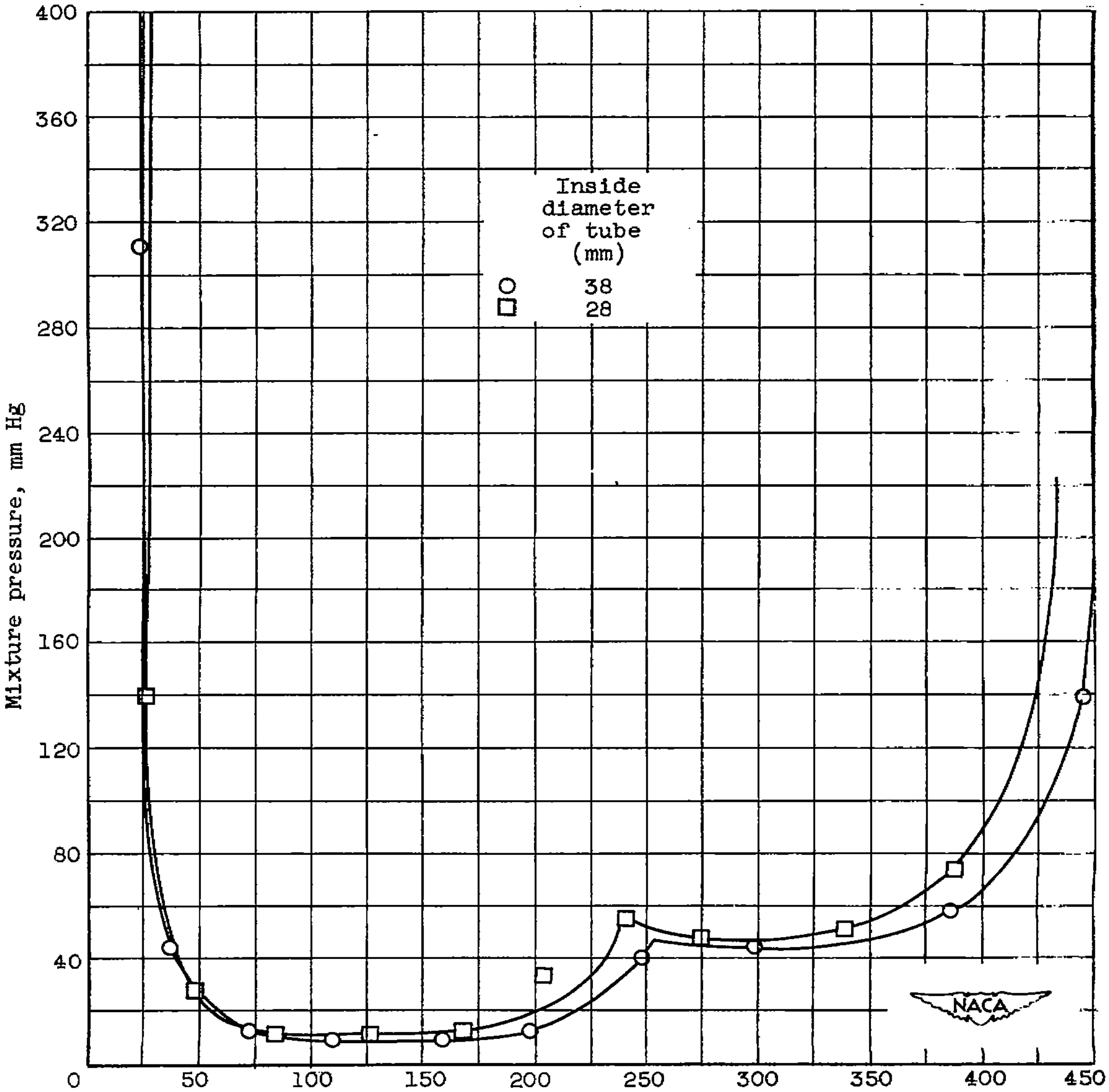


(e) Oxygen fraction $\frac{O_2}{O_2 + N_2}$, 0.347.

Figure 2. - Continued. Effect of tube diameter on pressure limit.



(f) Oxygen fraction $\frac{O_2}{O_2 + N_2}$, 0.496.

Figure 2. - Concluded. Effect of tube diameter on pressure limit.

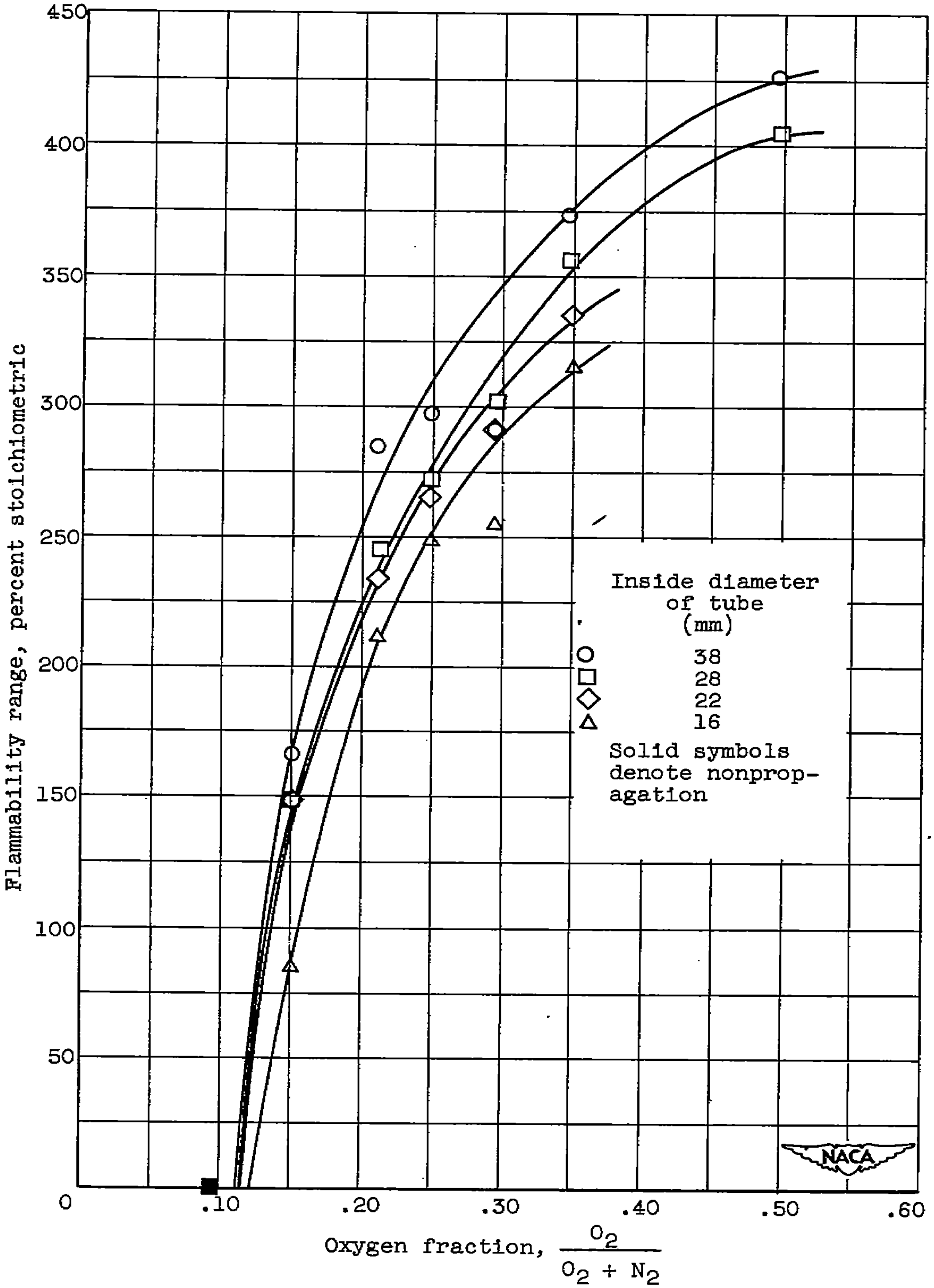


Figure 3. - Effect of oxygen on flammability range. Pressure, 250 millimeters of mercury.

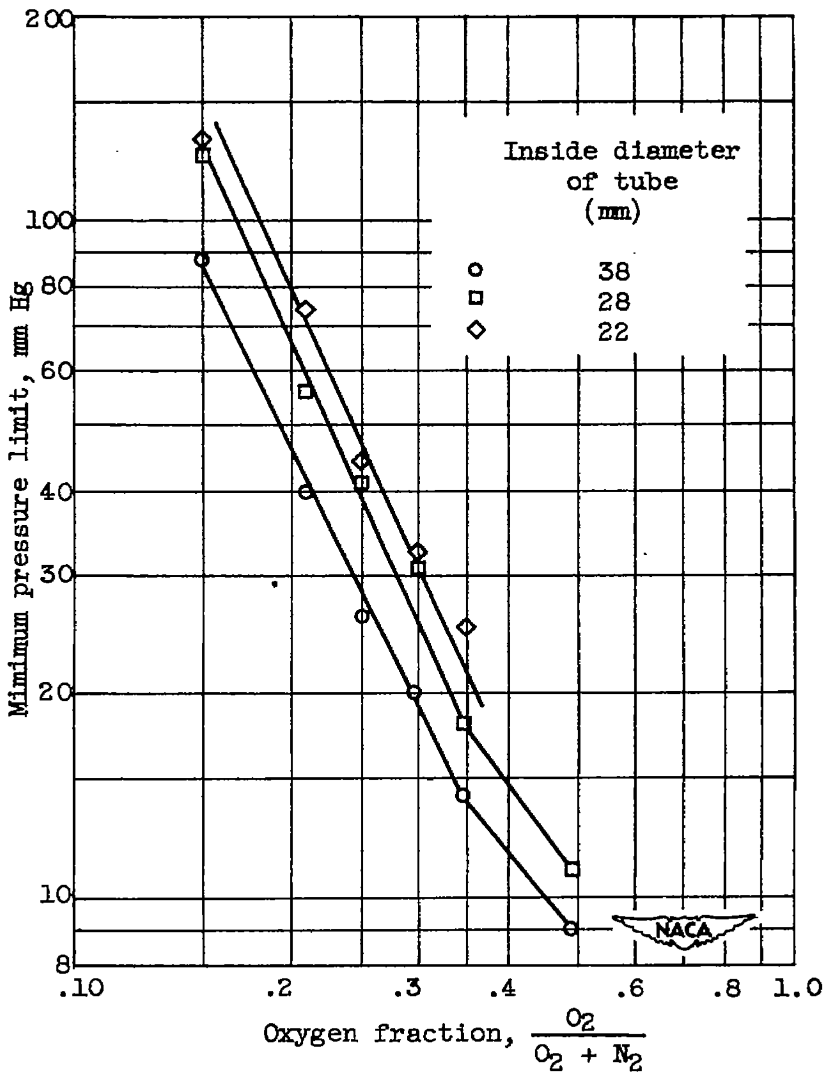


Figure 4. - Effect of oxygen on minimum pressure limit.

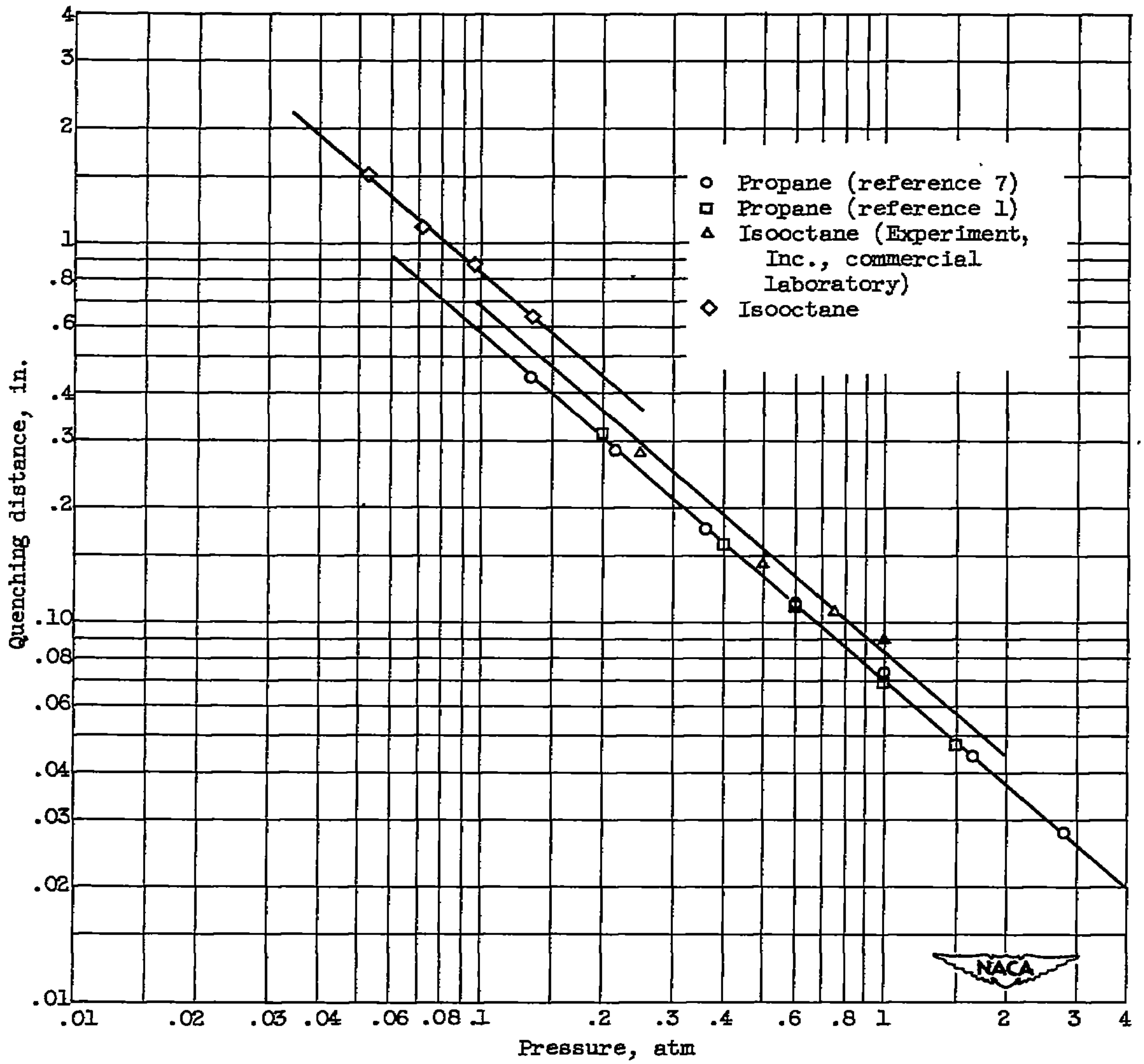


Figure 5. - Effect of pressure on quenching distance.

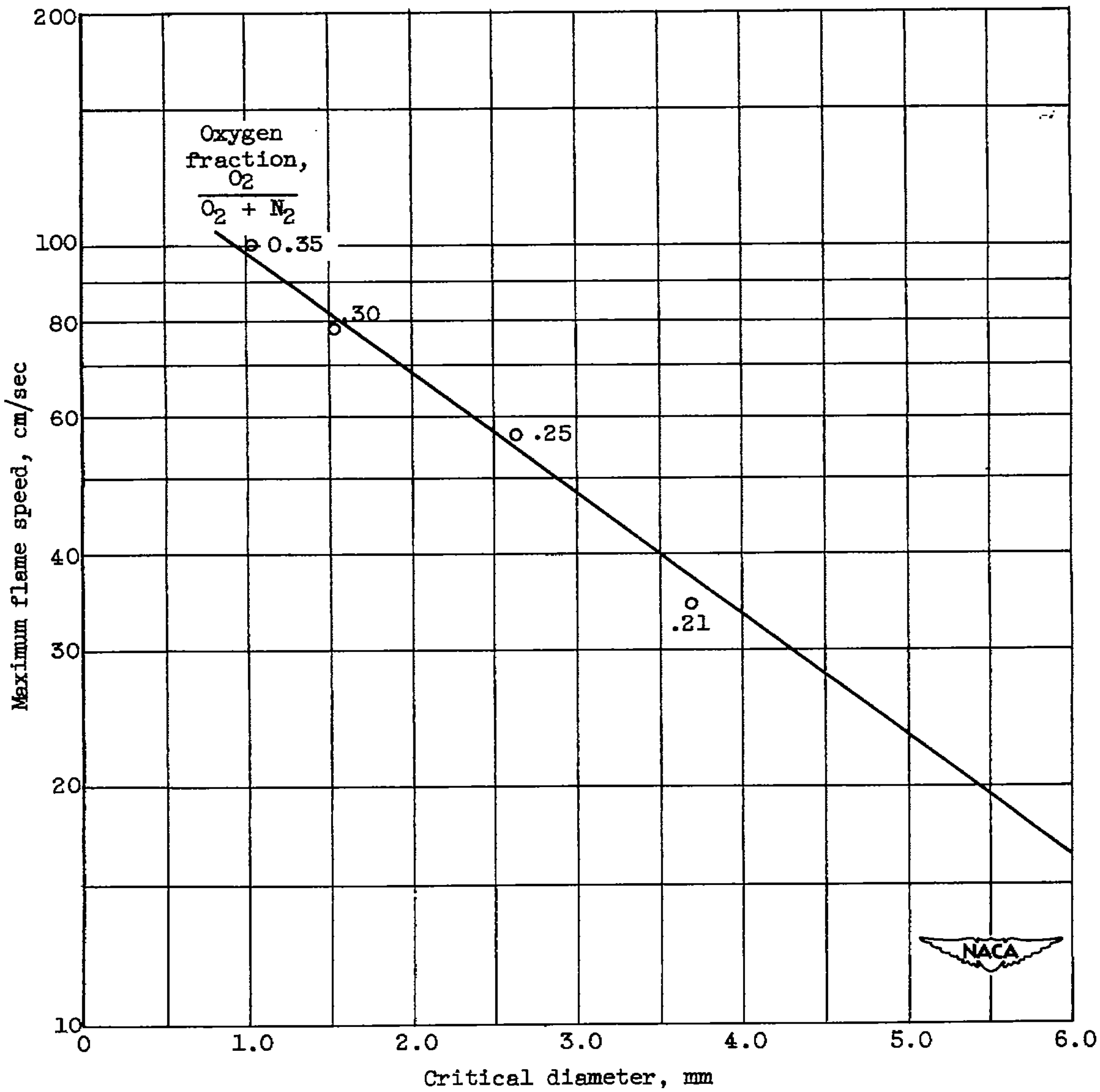


Figure 6. - Correlation of critical diameter with flame speed. Fuel, isooctane; pressure, 1 atmosphere; fuel-air ratio, 105 percent stoichiometric; temperature, 58° C.